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14. ABSTRACT

A narrow-band and widely tunable Optical Parametric Oscillator laser system has been acquired and installed during the performance period of this HBCU-Infrastructure award. The laser system has been incorporated in the development of a new high-resolution photoluminescence excitation setup. Initial spectroscopic studies of several novel photonic materials have been carried out (GaN:Er and GaN:Eu). The initial results have been presented at several conferences and a first manuscript of research results is in preparation.

The new instrumentation has been incorporated in the training of undergraduate and graduate students seeking B.S, M.S., and Ph.D. degrees in physics and related disciplines. In addition, the instrumentation has been implemented in research projects for students participating in summer outreach programs.

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HIGH RESOLUTION LASER SPECTROSCOPY OF NOVEL PHOTONIC MATERIALS

FINAL PERFORMANCE REPORT

Uwe Hömmerich

November 11, 2002

AFOSR-AF Office of Scientific Research

GRANT AWARD# F49620-01-0528

HAMPTON UNIVERSITY

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A. ACQUIRED EQUIPMENT

The following equipment has been acquired during the performance period of this award:

Spectra Physics
133 Terra Bella Ave
Mountain View, CA 94043

Optical Parametric Oscillator (OPO) laser system including:

- Nd: YAG Laser, 1750mJ, 10 Hz, PRO-270-10
- Enhanced Energy Option for PRO-270-10
- Beamlok-355 for PRO-270-10
- Standard linewidth MOPO
- Frequency doubler option for MOPO

Total cost: $188,490

Thorlabs
435 Route 26
Newton, NJ 07860

- Lightweight Optical Breadboard

Total cost: $1,284

B. SUMMARY OF MOST IMPORTANT RESEARCH RESULTS

The acquired instrumentation was delivered to Hampton University early spring and the initial installation was completed by the end of April 2002. Initial laser spectroscopic studies of new photonic materials have been carried out since the installation. The obtained results have been discussed at several conferences and resulted in one manuscript, which is in preparation for submission. Two graduate and undergraduate students have participated in experiments using the new instrumentation during the performance period of this award.

In the following the most important research results are briefly summarized:

1. Development of a high-resolution photoluminescence excitation (PLE) setup up

The acquisition of the new narrow-band Optical Parametric Oscillator (OPO) laser system has allowed the P.I. to develop a new time-resolved photoluminescence excitation setup. The new PLE setup is currently being tested for excitation studies in the visible spectral region. Improvements of the setup and the extension of the PLE studies to the UV and infrared spectral region are still ongoing. Initial spectroscopic studies have been carried out on several new photonic materials. In the following, a description of the new PLE setup is outlined.

Photoluminescence Excitation (PLE) is a powerful spectroscopic tool to identify and de-convolute absorption bands of emitting centers. In PLE the luminescence intensity of an optical center is recorded as a function of excitation wavelength. Strong emission at a particular excitation wavelength indicates that the emitting center absorbs strongly at that wavelength. In this way it is possible to determine shape and position of absorption bands, which lead to the emission process. Commonly, PLE studies in the visible spectral region are performed using high power tungsten or xenon lamps. However, these sources do not provide enough light output to study rare earth absorption bands in thin films with high spectral resolution.

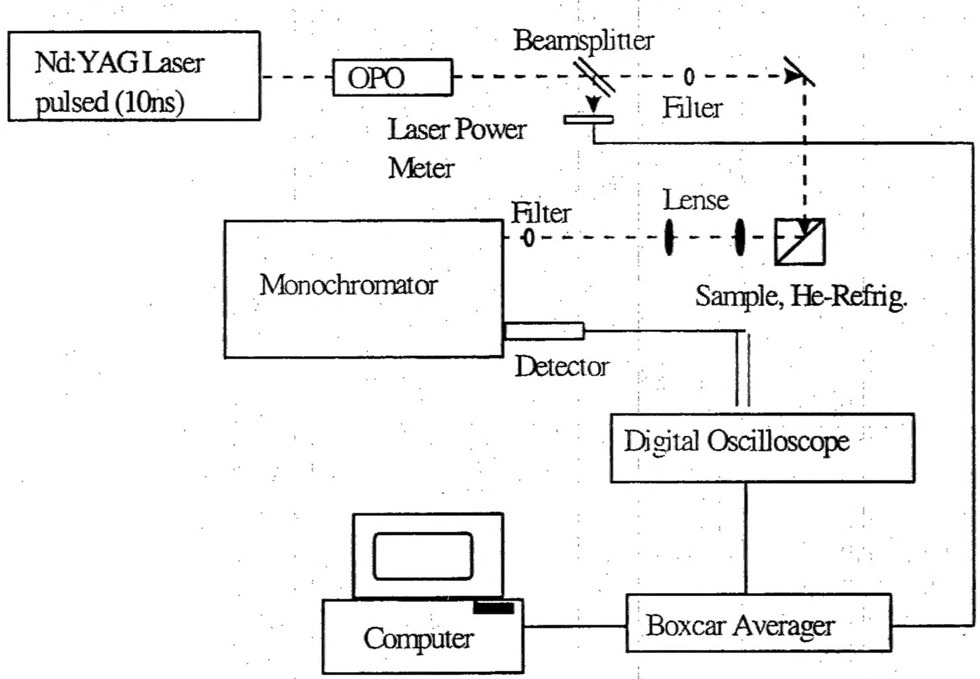

Figure 1: Experimental setup for time-resolved PLE measurements.

During the funding period of this HBCU instrumentation award, we have developed an experimental setup to carry out time-resolved photoluminescence excitation (PLE) studies over the wavelength range ~220-1800 nm. A schematic of this setup is shown in Figure 1. The key element of this setup is the newly acquired

narrow-band and tunable laser consisting of an optical parametric oscillator (OPO) pumped by a Q-switched Nd:YAG laser (5-10ns pulses). The resolution in the PLE studies is limited by the bandwidth of the OPO system, which is ~0.1-0.2 cm^{-1} allowing for high-resolution PLE measurements. During the PLE studies, the OPO output power is monitored using a pyroelectric power meter to normalize the PLE signal. The photoluminescence is dispersed with a single grating 0.5 m monochromator and detected with either a photomultiplier tube for visible emission studies or a LN cooled Ge detector for infrared studies. A long-pass filter is placed in front of the entrance slit of the monochromator to minimize stray laser light. A boxcar averager (time-resolved PLE experiments) is used to process the data. Temperature dependent PL measurements are conducted using a closed-cycle helium refrigerator for the range from 15-300 K.

In summary, a PLE setup was developed which consists of a narrow-band, pulsed laser system, several emission detectors, a gated boxcar averager, and a sample refrigeration unit. This PLE system allows one to carry out site-selective and time-resolved PLE measurements as a function of temperature (15-300 K). Further improvements of this PLE setup in terms of wavelengths coverage will be carried out in the future.

2. Laser spectroscopy of Er^{3+} doped GaN

The recent demonstration of visible (blue, green, red) and infrared (1.54 μm) electroluminescence from rare earth (RE) doped GaN has spurred significant interest in this class of materials for possible applications in optical communications and full color displays [1-4]. Compared to previously studies semiconductors such as GaAs or Si [2-4], GaN has some major advantages as a host for Er and other RE dopants. GaN is a wide-gap semiconductor, which leads to a reduced RE emission quenching and the observation of strong RE emission at room temperature. In addition, initial studies have shown that RE ions can be incorporated into GaN at concentrations as high as at least $1x10^{21}$ cm^{-3} [1,4]. In order to optimize the performance of current RE doped GaN devices it is important to obtain a better understanding of the RE incorporation, excitation schemes, and luminescence efficiency.

A sample of Er doped GaN prepared by solid-source MBE was provided by Dr. Steckl's group at the University of Cincinnati. The Er doped GaN sample prepared by solid-source molecular beam epitaxy (SSMBE) was grown in a Riber MBE-32 system on Si (111) substrates [5]. Ga and Er solid sources were used in conjunction with a rf plasma source supplying atomic nitrogen. The sample was pretreated by cleaning in acetone, methanol, and deionized water before insertion into the loadlock. The sample was subsequently outgassed at ~950°C before growth. During the growth, the Ga cell temperature was kept constant for a beam equivalent pressure of ~$8.2x10^{-7}$ torr. The rf-plasma source was kept constant at 400 W with a N_2 flow rate of 1.5 sccm, corresponding to a chamber pressure of mid -10^{-5} Torr. The growth temperature was varied from 750-950°C and the Er cell temperature was maintained at 1100°C. The Er concentration in the sample was determined to be ~$2x10^{20}$ cm^{-3}.

Figure 2 shows the visible PLE spectrum of Er doped GaN during MOMBE growth. This spectrum was recorded using the new instrumentation and PLE setup described before. The Er^{3+} PL was monitored at 1.535 μm, while varying the excitation wavelength from ~420 to 680 nm. Important information can be drawn from the PLE results. Striking features of the PLE spectrum are sharp peaks located at 495, 525, 553, and 651 nm. These sharp peaks coincide with the following Er^{3+} intra-4f transitions: $^4I_{15/2}\rightarrow{}^4F_{7/2}$, $^4I_{15/2}\rightarrow{}^2H_{11/2}$, $^4I_{15/2}\rightarrow{}^4S_{3/2}$, $^4I_{15/2}\rightarrow{}^4F_{9/2}$, $^4I_{15/2}\rightarrow{}^4I_{11/2}$, respectively, and are assigned to resonant Er^{3+} excitation. The sharp peaks seen in the PLE of Er: GaN are similar to absorption features of Er doped insulating materials, e.g. silicate based glass fibers. A higher resolution PLE spectrum of the $^4I_{15/2}\rightarrow{}^2H_{11/2}$ transition is shown in Figure 3 and compared to our previous PLE studies carry out with a broad band-excitation source. Clearly, the newly acquired narrow-band laser system allows a more detailed investigation of the energy levels of rare earth ions in GaN (and other thin-film hosts) and enables one to identify individual Stark-energy levels.

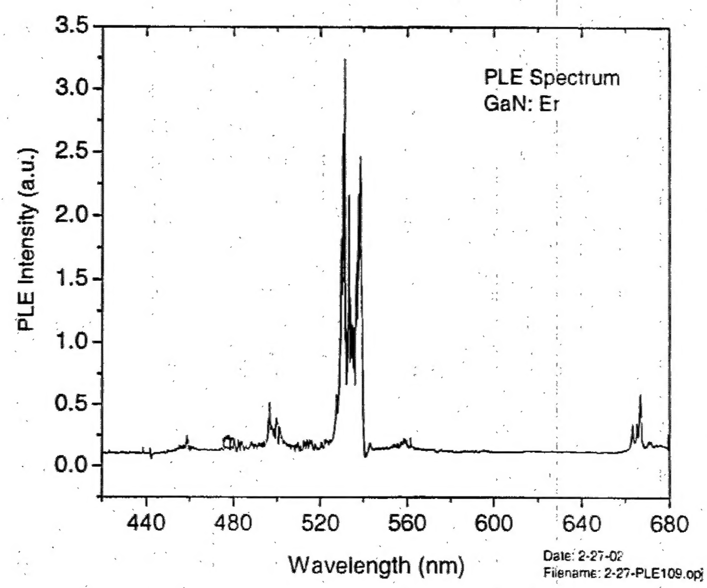

Figure 2: High-resolution ($\Delta\lambda$~0.1cm^{-1}) PLE spectrum of Er doped GaN prepared by SSMBE at room temperature.

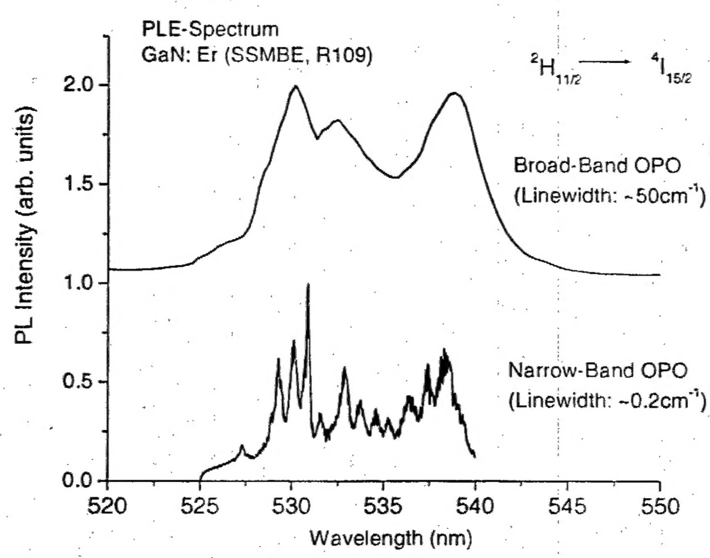

Figure 3: High resolution PLE spectra of the $^4I_{15/2} \rightarrow {}^2H_{11/2}$ transition of Er^{3+} ions in GaN. The upper trace shows the PLE spectrum obtained using a broad-band laser source. The lower trace shows the PLE spectrum using the new high-resolution PLE system developed under this HBCU instrumentation award. Clearly, more details on the energy level fine-structure of Er^{3+} can be obtained using the new narrow-band laser source. The crystal-field analysis of these data is in progress.

The information of the higher lying energy levels obtained from PLE measurements are important for gaining deeper insight the Er excitation schemes in GaN as well as the incorporation of Er ions in GaN. More detailed high-resolution PLE studies will be carried out in the future to identify the fine-structure of other Er intra-4f energy levels. The obtained data will then be used to carry out crystal-field calculations from which the local site symmetry of Er^{3+} ions in GaN can be derived.

3. Laser spectroscopy of Eu doped GaN (SSMBE)

The visible light emission from rare earth doped GaN has become of significant current interest for applications in display technology [1,4]. For obtaining red light emission, the $^5D_0 \rightarrow {}^7F_2$ transition of trivalent Eu ions seems most promising. Intense red line emission around 622 nm from Eu doped GaN has been demonstrated from several research groups [6-14] and EL devices have been fabricated [1]. The optimization of current devices requires, however, a more detailed understanding of the incorporation and emission properties of Eu^{3+} ions in GaN.

A sample of Eu doped GaN prepared by solid-source MBE was provided by Dr. Steckl's group at the University of Cincinnati. The Eu doped GaN film was grown in a Riber MBE-32 system on 2 inch p-Si (111) substrates [6]. Solid sources were used to supply the Ga (7N purity) and Eu (3N purity) fluxes. A RF plasma source was used to generate atomic nitrogen. For the nitrogen plasma an RF power of 400 W and an N_2 flow rate of 1.5 sccm was used. The Ga cell temperature ranged from 870 to 890C. A GaN buffer layer was first deposited for 10 min at a substrate temperature of 600C. For the main growth the substrate temperature was ramped to 800C. The Eu cell temperature was varied from 350 to 450C for the growth of various films. The Eu concentration is estimated to be $\sim 10^{18}$-$10^{21}/cm^3$.

An overview of the visible photoluminescence spectrum of the investigated Eu doped GaN sample at room-temperature is shown in Figure 4. The emission was excited using a HeCd laser. Only weak band-edge PL from the GaN host was observed at ~365 nm. Depending on the sample position, some weak yellow-band emission was observed extending from ~450-700 nm. Superimposed on this emission were intra-4f emission lines from Eu^{3+} ions located at ~545 nm ($^5D_1 \rightarrow {}^7F_1$), 601 nm ($^5D_0 \rightarrow {}^7F_1$), 623 nm ($^5D_0 \rightarrow {}^7F_2$), and 665 nm ($^5D_0 \rightarrow {}^7F_3$).

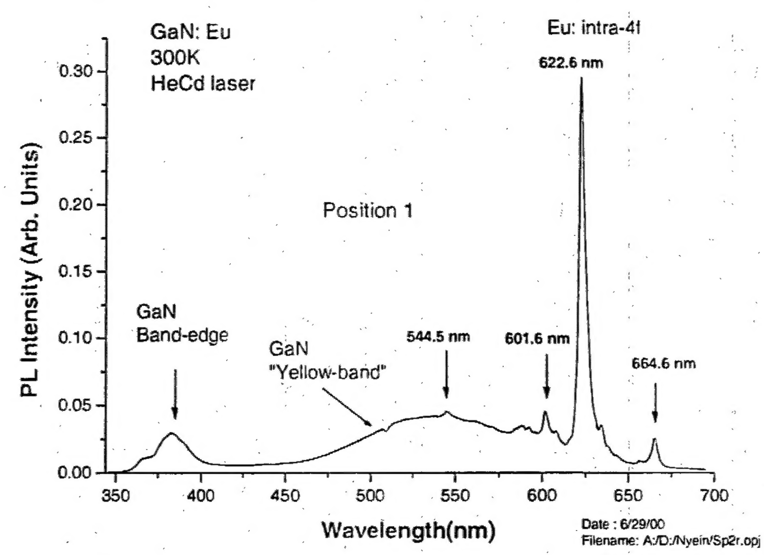

Figure 4: Overview of the visible emission from Eu doped GaN under above-gap excitation.

Excitation wavelength dependent emission studies were carried out on Eu doped GaN at 15 K using above-gap (carrier-mediated) and below-gap (resonant) excitation. Similar to Er doped GaN, the PL spectrum of Eu doped GaN changes slightly for different excitation wavelengths. In addition, the PL lifetime changes for the two different excitation wavelengths. This observation indicates that Eu^{3+} ions are incorporated into different sites in the GaN lattice. More site-selective PL studies are planned using the new narrow-band OPO system and will assist to identify the lattice sites of Eu^{3+} ions in GaN.

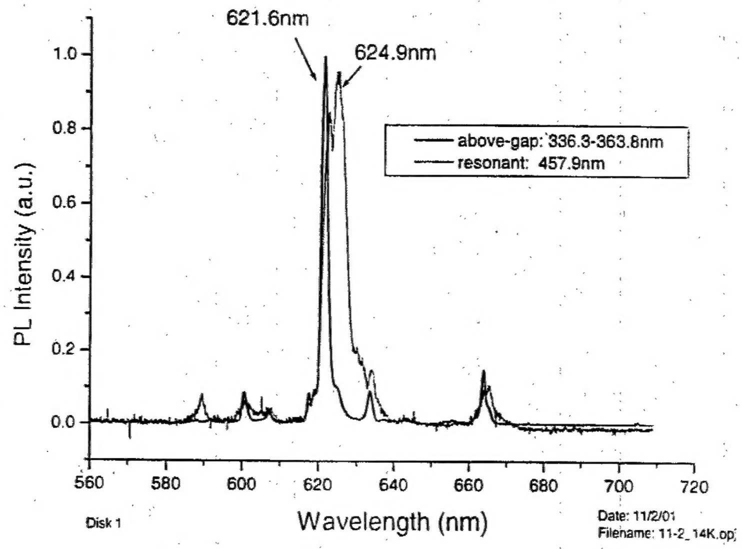

Figure 5: High resolution PL spectra of Eu doped GaN using above and below-gap excitation at 15K.

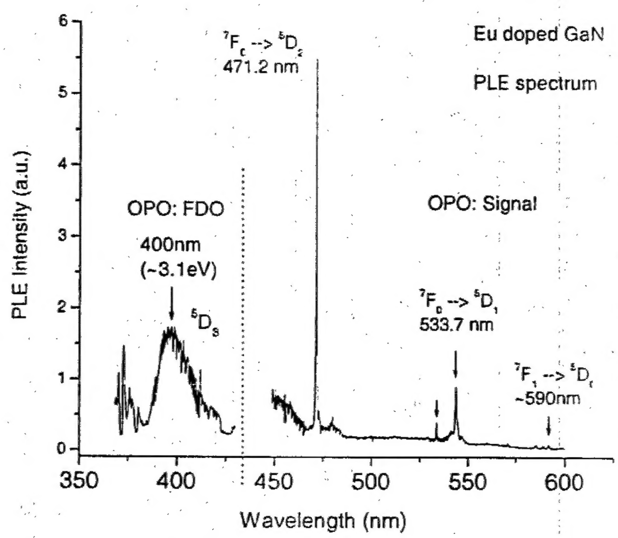

Figure 6: Initial PLE spectrum of Eu doped GaN at 300K.

An initial PLE spectrum of Eu doped GaN is depict in figure 6. Narrow intra-4f Eu^{3+} absorption lines can be identified at ~471.2 nm ($^7F0 \rightarrow {}^5D_2$); ~530-550nm ($^7F_0 \rightarrow {}^5D_1$), and at ~590 nm ($^7F_1 \rightarrow {}^5D_0$). In addition, a broad excitation band centered at ~ 400 nm (~3.1 eV) was observed, which overlap some higher lying Eu^{3+} levels ($^7F_0 \rightarrow {}^5D_3$, $^7F_0 \rightarrow {}^5L_6$). Based on FTIR measurements, Bang et al. [11,12] recently reported a Eu-related defect level at 365 meV below the conduction-band of GaN. A defect-related energy transfer model had been proposed in the past by several authors for the excitation of Yb^{3+} in InP and subsequently also for Er^{3+} for III-V's and Si [2-4]. A broad PLE excitation band was also reported for Er implanted GaN [15]. These results suggests that for an efficient excitation of rare earth ions in GaN, an overlap of higher lying RE excited states with the RE-related defect energy is necessary in order to facilitate an efficient carrier-mediated excitation process. More comparative PLE studies of other rare earth doped GaN systems are currently in progress to further support this excitation model.

C. EDUCATIONAL IMPACT

Undergraduate and graduate students have participated in the installation of the new OPO laser system as well as the development and testing of a new high-resolution PL and PL excitation setup. One undergraduate student from engineering (Mr. Osei Poku) has been instrumental in developing a nitrogen purge system for the new OPO laser system. A physics undergraduate student (Ms. Brandi Cade, figure 7) has assisted during a summer outreach program in developing and testing a new PL and PLE setup. Two graduate students (Ms. Althea Bluiett and Ms. Ei Ei Nyein, figure 8) are currently running PL and PLE excitation studies on RE doped GaN and solid-state laser materials.

In summary, the new instrumentation has been incorporated in the training of undergraduate and graduate students seeking B.S, M.S., and Ph.D. degrees in physics and related disciplines. In addition, the instrumentation has been implemented in research projects for students participating in summer outreach programs. In the future, students and faculty from other science and engineering departments at H.U. will be trained in the use of the new laser systems and collaborative research projects will be developed.

Figure 7: Brandi Cade, a physics undergraduate student at H.U. aligning lenses and mirrors for a new PL setup.

Figure 8: Ei Ei Nyein and Althea Bluiett, graduate students in the department of physics at H.U., aligning a laser experiments. Both students carried out several PL experiments using the new instrumentation acquired through this grant. Ms. Bluiett is near completion of her Ph.D. in physics and was recently awarded a NRC postdoctoral fellowship at the Navy Research Laboratory in Washington.

D. LIST OF CONFERENCE PRESENTATIONS AND PUBLICATIONS

a) Conference Presentations

U. Hömmerich, J. Zavada, A. J. Steckl, "Luminescence Properties of Rare Earth doped GaN", 23rd Rare Earth Research Conference, Davis, CA, July 13-18, 2002, invited talk.

Ei Ei Nyein, U. Hömmerich, J. Heikenfeld, D. S. Lee, A. Steckl, J. M. Zavada, "Spectroscopic evaluation of rare earth doped GaN for full-color display applications", Conference on Lasers and Electro-Optics/Quantum Electronics & Laser Science (CLEO/QELS) Long Beach, CA, May 19-24, 2002, Oral presentation, CFG3

Ei Ei Nyein, U. Hommerich, D. S. Lee, A. Steckl, J. M. Zavada, "Emission Properties of Er-doped GaN as a function of Ga flux, American Physical Society March 2002 Meeting, Indianapolis, Indiana, March 18-22, 2002, oral presentation, paper T18-6.

Ei Ei Nyein, J. T. Seo, U. Hommerich, "Evaluation of Eu doped GaN for phosphor applications", presented at the Virginia Academy of Science Meeting, May 23-25, 2001, Harrisonburg, VA.

b) Publications

Ei Ei Nyein, U. Hommerich, D. S. Lee, J. Heikenfeld, J.M. Zavada, Photoluminescence and Photoluminescence Excitation Studies of Eu doped GaN, in preparation for submission to Applied Physics Letters.

E. BIBLIOGRAPHY

[1] A.J. Steckl and J.M. Zavada, MRS Bulletin, 24(9), 33 (1999).

[2] G. S. Pomrenke, P.B. Klein, D. W. Langer, (Eds.), *Rare Earth Doped Semiconductors I*, Materials Research Society, Vol. 301, 1993.

[3] S. Coffa, A. Polman, and R. N. Schwartz, (Eds.), *Rare Earth Doped Semiconductors II*, Materials Research Society, Vol. 422, 1996.

[4] J.M. Zavada, T. Gregorkiewicz, and A. J. Steckl, (Eds.), *Rare Earth Doped Semiconductors III*, Materials Science & Engineering B, Vol. 81, 2001.

[5] A.J. Steckl and R. Birkhahn, Appl. Phys. Lett. 73 (1998) 1700.

[6] J. Heikenfeld, M. Garter, D.S. Lee, R. Birkhahn, and A. J. Steckl, Appl. Phys. Lett. **75**, 1189 (1999).

[7] S. Morishima, T. Maruyama, M. Tanaka, Y. Masumoto, K. Akimoto, Phys. Stat. Sol. **A 176**, 113 (1999).

[8] H. J. Lozykowski, W. M. Jadwisienczak, J. Han, I.G. Brown, Appl. Phys. Lett. **77**, 767 (2000).

[9] T. Monteiro, C. Boemare, M. J. Soares, R. A. Sa Ferreira, L. D. Carlos, K. Lorenz, R. Vianden, E. Alves, Physica B **308-310,** 22 (2001).

[10] M. Overberg, K.N. Lee, C. R. Abernathy, S. J. Pearton, W. S. Hobson, R. G. Wilson, J. M. Zavada, Materials Science and Engineering **B81** 150 (2001).

[11] H. Bang, S. Morishima, Z. Li, K. Akimoto, M. Nomura, and E. Yagi, Phys. Stat. Sol. (b) **228**, 319 (2001).

[12] H. Bang, S. Morishima, Z. Li, K. Akimoto, M. Nomura, E. Yagi, J. Crystal Growth **237-239,** 1027 (2002).

[13] Z. Li, H. Bang, G. Piao, J. Sawahata, K. Akimoto, J. Crystal Growth **240**, 382 (2002).

[14] Ei Ei Nyein, U. Hömmerich, J. Heikenfeld, D. S. Lee, A. J. Steckl, J. M. Zavada, "Spectroscopic evaluation of rare earth doped GaN for full color display applications", in OSA *Trends in Optics and Photonics (TOPS) Vol. 73, Conference on Lasers and Electro-Optics*, OSA Technical Digest, Postconference Edition (Optical Society of America, Washington, DC, 2002), p 654.

[15] S. Kim, S. J. Rhee, X. Li, J. J. Coleman, S. G. Bishop, and P. B. Klein, J. Eletron. Mater. **27**, 246 (1998).